Programmable Digital Droplet Microfluidics Using a Multibarrel Capillary Bundle

Renee R. Hood,^{†§} Thomas Wyderko,[†] and Don L. DeVoe^{†§*}

[†]Department of Mechanical Engineering

[§]Fischell Department of Bioengineering

University of Maryland

College Park, MD 20742

*Corresponding author: Don DeVoe

Email: ddev@umd.edu

Phone: 301-405-8125

Fax: 301-405-8125

Abstract

The integration of multibarrel glass capillaries into a microfluidic co-flow configuration is demonstrated for parallel production of nanoliter scale droplets. The multibarrel system supports the generation of a continuous stream of single-phase or dual-phase droplets with digital control over droplet content, enabling the formation of dynamically-programmable and tunable emulsions. The multibarrel capillary method allows simultaneous generation of multiple droplets from parallel droplet generators, with controlled switching between selected dispersed phase or continuous phase compositions. Individual phases emerging from each capillary outlet may be selected to yield merged droplets combining continuous phase emitted from one or more neighboring droplet generators. Because the capillary emitters are configured in a two dimensional array, the multibarrel system provides greater flexibility toward producing controlled interactions between multiple solutions and phases than conventional planar microfluidic droplet generators. Using a 7-barrel capillary configuration, the ability to regulate droplet composition and particle size of individual as well as combined single and merged water-in-oil droplets is explored, together with the use of two-dimensional spatial control within the multibarrel array to generate higher order emulsions.

Keywords: Emulsions; Multibarrel pipettes; Microparticles; Capillary bundle

1. Introduction

Emulsions are systems comprising droplets of a compartmentalized fluid dispersed within a second immiscible continuous phase. With the ability to form droplets with diameters ranging from the nanoscale to the microscale, emulsions play a large range of roles for applications including encapsulation of food additives [1,2], targeted drug delivery [3–7], special material processing [8,9], and various other technological fields [10,11]. Nearly all applications of emulsions depend strongly on the suspended droplet properties, such as droplet composition, size, and size uniformity [12]. Numerous emulsification methods have been developed at the bulk scale, with the majority involving mixing of two immiscible liquids within turbulent flows to promote top-down droplet formation. Emulsification processes within this category offer limited control over individual particle formation, resulting in populations of relatively large droplets with broad size distributions [13]. Alternative methods enabling bottom-up droplet formation leveraging microfluidic flows have been widely investigated, with exceptional control over particle generation within laminar flow fields resulting in highly uniform emulsions [12–17]. Microfluidic synthesis methods generally yield droplets with diameters ranging from a few micrometers to hundreds of micrometers formed within a uniform and continuous stream [12]. In addition, microfluidic techniques enable the creation of multiphase particles [18], anisotropic particles [19], and other novel particle types [20] where droplet size and the number of species encapsulated within each droplet can be manipulated with unprecedented accuracy [21–24].

While microfluidic-based droplet synthesis techniques have proven highly successful for the formation of a wide range of emulsions, platforms based on these methods are typically configured as static systems that support the formation of specific emulsion types with predefined droplet content. Although droplet composition in these systems may be adjusted by changing the dispersed phase introduced into the device, for example by using off-chip selection valves to switch between different input fluids, this approach requires that the entire fluidic network be flushed between each droplet formation state, limiting the speed and flexibility of the system. An alternate approach to controlling droplet makeup is to form multiple droplets containing different phases or chemical species using individual droplet generators, followed by selective merging of the droplets to achieve the final desired composition [25,26]. However, this approach remains limited in its ability to dynamically adjust the individual droplet content prior to merging, and does not readily support higher levels of multiplexing. More fundamentally,

because each droplet emitter within a planar microfluidic droplet generator can have at most two neighboring emitters, the ability to combine multiple dispersed phase components within small individual droplets is limited. Overall, established microfluidic systems do not provide effective means for dynamically generating multiplexed droplets for the formation of customizable and dynamically tunable emulsions.

Multibarrel glass capillaries are comprised of large-diameter pipette bundles that are heated and pulled to reduce the diameter and fuse the bundle into a single element. Although most commonly used as microelectrode arrays for high resolution electrophysiology measurements, these capillary bundles have recently utilized for the production of nanoscale liposomes [27] as well as microscale double emulsions [28]. The parallel flow paths and precise positioning of the fluid emitters that can be achieved using multibarrel capillaries present new opportunities for multiplexed droplet generation. In particular, multibarrel capillaries possess outlet microchannels which are configured in a two dimensional array. As a result, when using a multibarrel emitter for microfluidic droplet generation, the platform provides a means for controlling interactions between large numbers of adjacent flow paths, and thus provides a significantly greater level of flexibility when compared to single-barrel co-flow systems or planar microfluidic droplet generators. Here we demonstrate that by interfacing each inlet within the multibarrel capillary to a shutoff valve, individual capillaries within the bundle may be connected to discrete fluid species and independently controlled for the introduction of multiple dispersed and continuous phases within the flow system. This unique integrated format enables a number of different single (unary) and merged (binary) droplet combinations to be produced in a continuous-flow manner.

The capillary-based device explored here consists of a pulled 7-barrel glass capillary connected to additional tubing for supply of fluids (Fig. 1). Although similar to traditional co-flow microfluidic droplet generators in terms of the flow physics, this unique configuration allows the interaction of multiple fluid streams within the co-flow system, enabling higher order droplet formation and a variety of possible emulsion combinations that can be digitally controlled for both composition and size by opening or closing individual fluid inlets. The inlet ends of the multibarrel capillaries are connected to oil and water supply lines, and the pulled end of the multibarrel capillary is encased by a glass micropipette to support sheath flow of a continuous oil phase surrounding the dispersed phase droplets emerging from the multibarrel

array. The resulting apparatus is a low pressure system assembled using inexpensive components, requiring no costly equipment, infrastructure, or fabrication processes.

2. Materials and methods

2.1. Multibarrel capillary device assembly

Microfluidic capillary devices were fabricated from an assembled combination of a multibarrel glass capillary and various tubing. Each capillary of the 7-barrel pipette (FHC, Inc., Bowdoinham, ME) was interfaced with PEEK tubing (Upchurch Scientific, Oak Harbor, WA) and Tygon tubing (Cole Parmer, Vernon Hills, IL) for supply of dispersed (water) and continuous (oil) phases to assist in droplet production. Four of the seven capillaries were connected to low pressure micro-splitter valves (Upchurch Scientific) to enable dynamic switching between continuous and dispersed phases. The opposite end of the multibarrel capillary was pulled to a total tip diameter of 100 μm , and positioned within a 25 μL glass microcapillary (Microcaps, Drummond Scientific, Broomall, PA) with an approximate inner diameter of 700 μm . A glass Pasteur pipette was used to align the microcapillary to the multibarrel capillary as well as to hold the apparatus together.

2.2. Reagent preparation

The continuous phase oil solution was comprised of corn oil (Mazola, ACH Food Companies, Inc., Cordova, TN) with Span 80 (2 vol %) (Sigma Chemical Co., St. Louis, MO) as a surfactant. The dispersed phase aqueous solution contained deionized water with various colors of food dye to aid in visualization.

2.3. Droplet generation

All fluids were delivered through programmable syringe pumps (PHD2000, Harvard Apparatus, Holliston, MA). For droplet formation, if the oil phase was connected through the switch valve, the linear velocity was constant at $V_{\text{oil}} = 0.34 \text{ cm/s}$ at the pulled capillary exit (corresponding to $Q_{\text{oil}} = 0.18 \text{ }\mu\text{L/min}$) to match the overall sheath flow linear velocity ($Q_{\text{oil}} = 100 \text{ }\mu\text{L/min}$). When the water phase was connected through the valve, hence the inlet was forming two-phase or single droplets, the velocity remained constant at $V_{\text{water}} = 1.53 \text{ cm/s}$ ($Q_{\text{water}} = 0.8 \text{ }\mu\text{L/min}$) unless otherwise noted. Images of the droplet generation process and

collected droplets in Figures 3, 5, and 6 were modified for clarity using background subtraction and contrast enhancement to aid the reader in identifying specific droplet populations.

2.4. Droplet characterization

Populations of droplets were collected at the outlet of the device and transferred to standard glass microscope slides for characterization. The droplets were visualized using a Nikon Eclipse LV-100 microscope (Nikon, Melville, NY) and measured manually for diameter using NIS Elements software (Nikon). At least 35 droplets from each population were measured, with each individual droplet measured 3 times along different axes, to obtain the size distribution. A normal distribution was assumed for plotting the cumulative sizes of each population of droplets.

3. Results and discussion

As an initial study to evaluate this approach, multiple inlets to the multibarrel capillary device were connected to oil and water phases through micro-splitter valves, allowing the selected inlets to be dynamically switched between the dispersed aqueous phase and the continuous oil phase. When adjacent capillaries are both configured to emit dispersed phase, the developing droplets merge, thereby enabling the multibarrel output to alternate between single droplet and dual droplet formation through digital control of the inlet valves (Fig. 2). When forming individual droplets, the process is asynchronous, since there is no direct coupling between the individual droplet generators. However, for the case of merged droplets, coupling between the fluid volumes ensures that individual merged droplets are emitted from the paired capillary outlets, with no satellite droplets observed. While consistent droplet merging and uniform binary droplet volume were achieved when using identical flow rates for each of the dispersed phases, merged droplets may also be generated when using different mixing ratios. However, because merging of droplets from individual emitters requires that the dispersed phase fluids come into contact prior to droplet pinch-off, the flow regimes within which consistent droplet merging occurs is dependent on individual dispersed phase flow rates, continuous phase flow rate, and resulting droplet volumes. Understanding the impact of these parameters on the control and uniformity of binary droplet production, and identifying regimes of stable droplet merging, will require further investigation.

Due to dead volume and compliance within the capillaries and connecting tubing, reaching steady state following each switching event was found to require approximately 20 s for the device configuration and flow rates used in this experiment.

The multibarrel capillary device allows for individual inlets to produce discrete droplets when connected to aqueous dispersed phase inlets (Fig. 3a, Supplemental Video 1) as well as multi-compartment droplets when adjacent capillaries are connected to the aqueous dispersed phase, thus allowing coalescence of the 2 fluids before the droplet is pinched off by the surrounding oil continuous phase (Fig. 3b, Supplemental Video 2). This feature may be particularly useful in emulsion systems with complex requirements, such as unique combinations of droplets, droplets containing multiple compartments, or droplets in which well-controlled chemical reactions between multiple chemical species contained within discrete fluids are desired. Here we demonstrate the device's unique ability to produce customizable emulsion systems by connecting 4 of the 7 capillaries in a bundle to micro-splitter valves for digital flow control, enabling these inlets to dynamically alternate between producing a single or two-phase droplet to producing continuous phase sheath flow without the need for flow disruption or device reassembly.

Before evaluating the performance of this multibarrel configuration for multi-droplet formation, the size distribution of populations of single droplets forming from each individual inlet was determined (Fig. 4). The flow velocity of the dispersed aqueous phase was held constant at 1.53 cm/s for each droplet configuration. For each test, all capillaries other than the selected dispersed phase capillary were connected to the continuous oil phase, each with a flow rate chosen to match the average linear flow velocity for continuous phase within the sheath flow surrounding the overall capillary bundle (0.34 cm/s). The average diameter of the resulting droplets was 203 μm , with a relative standard deviation of 2.3% observed between droplets produced by different dispersed phase inlets within the capillary bundle. For the flow rates employed in this experiment, the droplet production frequency was approximately 1.0 droplets/s for each dispersed phase inlet.

A significant advantage of microfluidic systems for production of emulsions is the ability to manipulate the diameters of the resulting droplets by simply varying flow conditions. The capacity of the multibarrel capillary device to independently control individual droplet size by varying the flow rate of the dispersed aqueous phase was investigated by holding the continuous

phase average flow velocity constant at 0.34 cm/s while varying the dispersed phase velocity between discrete values of 0.38 cm/s, 1.53 cm/s, and 38.2 cm/s, resulting in a dispersed: continuous flow velocity ratio of spanning two orders of magnitude from 1.1 to 110. The diameters of the corresponding droplets produced under these flow conditions were directly correlated with flow rate ratio (FRR), defined as the ratio of linear flow velocity of the dispersed phase to the linear flow velocity of the continuous phase. Over the selected range of FRR, droplet the average diameter varied between approximately 100 μm and 450 μm , as displayed in Fig. 5. Droplet generation rates of approximately 4.9, 2.3, and 5.2 droplets/s observed for dispersed phase velocities of 0.38, 1.53, and 38.2 cm/s, respectively.

The multibarrel system enables dispersed phase to be ejected from two or more adjacent capillary outlets, displacing oil between the emerging droplets and allowing the multiple aqueous solutions to merge before separating from the capillary array. In this work we explored the formation of paired droplets from two adjacent capillary outlets by imposing equal flow rates for aqueous dye solutions within two adjacent dispersed phase capillaries. Flow rates were selected to form combined droplets with diameters ranging from 215 μm to 265 μm , reflecting an approximate doubling of the average droplet volume between these cases. Over this range of droplet size, droplet merging was highly repeatable, with no missed merging events after reaching steady state. Furthermore, switching between single and dual droplet generation was investigated for all possible inlet configurations for the device described in Fig. 2. The size distribution of each single and dual droplet formed within the multiplexed system was compared to the size distribution when formed alone to assess the effect of adjacent droplet formation on particle size (Fig. 6). As in the initial experiments with droplet formation from individual capillaries, the average flow velocity for the continuous phase within both the capillary and sheath flows was held constant at 0.34 cm/s, with the dispersed phase velocity varied to achieve the desired droplet volumes. In all cases, stable parallel formation of single and dual droplets was achieved, with successful switching between stable states within approximately 20 s using the external valves. As seen in Fig. 6, when simultaneously forming single and dual droplets, the resulting droplet sizes varied from those observed when forming single or dual droplets alone. The observed changes likely reflect hydrodynamic coupling between the parallel flow paths, and highlight the need for careful calibration of flow velocities and control over device symmetry during both capillary pulling and multibarrel assembly.

In principle, the simultaneous formation and merging of droplets described in this work can also be performed using conventional planar microfluidic droplet generators through the fabrication of multiple adjacent droplet emitters. However, an important aspect of the multibarrel capillary co-flow system is the ability to control merging between adjacent droplets arranged in a two-dimensional pattern, unlike planar microfluidic devices which are limited to one-dimensional arrangements of droplet emitters. By packing the droplet emitters into a two-dimensional configuration, multibarrel devices can support the merging of droplets from a significantly greater number of individual emitters, in a manner that is scalable through the use of higher density capillary bundles. Although not studied at the same level of details as the simultaneous unary/binary droplet generation described in Fig. 6, droplet merging is readily achieved for all combinations of outlets for the case of the 7-barrel capillary used in the present work. Furthermore, beyond simply increasing the number of droplets which can be merged, the ability to manipulate the spatial arrangement of the various fluid phases enables higher level control over the morphology of the resulting droplets. For example, by taking advantage of the two-dimensional spatial control of droplet emission afforded by the multibarrel system, higher order emulsions are possible [28]. By establishing a water (or oil) flow within each of the outer capillaries and an oil (or water) flow within both the inner capillary and outer sheath flow, merging of the phase emerging from the 6 outer capillaries occurs, and finally resulting in pinch-off of both phases for the formation of a double emulsion. Fig. 7 depicts a representative case where water/oil/water double emulsions were generated within the multibarrel system when using linear flow velocities of the inner, middle, and outer phases set to 1.34 cm/s, 0.32 cm/s, and 0.34 cm/s, respectively. For these particular flow velocities, the resulting emulsions exhibited inner droplet diameters of 57 μm and outer diameters of 251 μm . By adjusting the relative flow rates of the individual phases, the multibarrel device was also found to be capable of switching from the formation of highly uniform double emulsions to alternating single and double emulsions (Supplemental Videos 3 and 4).

4. Conclusions

Here we have demonstrated multiplexed droplet formation utilizing a pulled multibarrel capillary device with the ability to generate a continuous stream of customizable droplets. While further efforts are needed to investigate the impact of hydrodynamic coupling between multiple flow

paths within the system, the 7-capillary system explored in this work demonstrates the ability to adjust droplet size together with droplet composition through controlled merging of adjacent flow paths with simple digital control. Compared to conventional single barrel co-flow or planar microfluidic droplet generators, the multibarrel capillary device enables greater control over the simultaneous formation and merging of droplets containing discrete fluid species, allowing rapid digital tuning of droplet composition without the need to disrupt fluid flow or reconfigure the device. The resulting system presents unique opportunities for the multiplexed formation of complex and dynamically tunable emulsions. Beyond its initial demonstration in this work, the platform may be further extended by scaling to higher capillary array densities, and the multibarrel system may offer additional utility for the formation of anisotropic particles and other novel materials with controllable morphology and composition by combining digital droplet generation with in situ polymerization.

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Figures

Fig. 1. (a) Schematic of the pulled multibarrel capillary device with oil and water input tubing, showing the switch valve connections to 4 of the 7 capillary inlets, (b) magnified view of the pulled end of the capillary bundle, and (c) image of an assembled device.

Fig. 2. Configuration of inlets for the following experiments. The 7-barrel capillary contains 3 center inlets which are connected to continuous (oil) phase only and two sets of adjacent capillaries connected to continuous (oil) and dispersed (A,B,C,D; all water) phases separated by a valve, which enables rapid switching between phases for digital control over droplet formation for programmable emulsions.

Fig. 3. Schematic sequences illustrating droplet formation within the multibarrel capillary device: (a) single droplet formation with one capillary connected to an aqueous phase and all other capillaries and sheath flow connected to oil, and (b) dual droplet formation with two adjacent capillaries connected to aqueous phases which merge to form a single droplet of two combined fluids.

Fig. 4. Size distributions of droplets generated from individual capillaries with the multibarrel device, with a single inlet connected to dispersed aqueous phase at a time as noted (see Fig. 2 for inlet configuration). Similar droplet dimensions were observed when employing identical flow rates for each capillary, with only slight variations in droplet diameters which likely result from minor capillary asymmetries or error during manual device alignment.

Fig. 5. Size distributions for droplets formed using a single capillary within the multibarrel system as a function of flow rate ratio (FRR), defined as the ratio of dispersed:continuous volumetric flow rates.

Fig. 6. Size distributions of unary and binary droplets formed simultaneously (solid curves), compared to identical droplet types when formed alone (dashed curves) using the 7-barrel capillary device (Fig. 1). Each plot represents data from 3 specific inlet configurations with either unary droplet generation (X), binary droplet generation (YZ), or simultaneous unary and binary droplet generation (X+YZ), as indicated. Corresponding images of simultaneously formed unary and binary droplets are shown in the rightmost panels for each case.

Fig. 7. (a) Inlet configuration for the generation of water/oil/water double emulsions through the merging of fluid from 6 oil outlets surrounding a single aqueous outlet, with the oil streams themselves sheathes by water, and (b) resulting double emulsion droplets generated using the multibarrel capillary bundle (Fig. 1) with inner diameter $57.1 \pm 2.5 \mu\text{m}$ and outer droplet diameter $250.9 \pm 6.3 \mu\text{m}$.

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Renee R. Hood received her Ph.D. from the the Fischell Department of Bioengineering at the University of Maryland, College Park, in 2014, with an emphasis on microfluidic synthesis of liposomal nanomedicines. She is presently a Postdoctoral Fellow with the National Cancer Institute at the National Institutes of Health focusing on the development of microscale platforms for synthetic biology.

Thomas Wyderko is currently an undergraduate student at the University of Maryland, College Park with a dual major in Mechanical Engineering and Computer Science. He also currently works as a consultant for VMware Inc.

Don L. DeVoe is a Professor in the Department of Mechanical Engineering at the University of Maryland, with an affiliate appointment in the Fischell Department of Bioengineering. He received his Ph.D. in Mechanical Engineering from U.C. Berkeley in 1997, with a focus in piezoelectric MEMS. His current research interests include microfluidic bioanalytical technologies and piezoelectric microsystems.